NRL Report 6662

Second Harmonic Generation by Reflection From GaAs

James B. Langworthy
Nuclear Physics Division

January 19, 1968

NAVAL RESEARCH LABORATORY
Washington, D.C.

51

CONTENTS

ABSTRACT

The intensity of second harmonic light generated by reflection from a Gallium Arsenide crystal is calculated in detail in four cases of polarization. Relevant properties of GaAs are stated and an experiment is described. Based on this, constitutive relations are written in the weak field approximation and the second and third order material tensors are reduced using crystal symmetry. The nonlinear wave equation is solved in the parametric approximation when phase matching is not present. The boundary value problems for the fundamental entering the medium and for the second harmonic leaving are solved. The only significant null solution is found to be the Brewster's angle extinction. Some numerical exploration of the exact effects of absorption at both fundamental and second harmonic are made. They are found to be small and are negligible for qualitative purposes unless the imaginary part of the dielectric constant exceeds the real part. Variation of intensity with crystal orientation is described in detail. The occurrence of phase matching as a null solution of the wave equation is seen to offer a natural approach to the problem of obtaining the geometry of phase matching for the most general optical medium.

PROBLEM STATUS

An interim report on one phase of the problem; work is continuing.

AUTHORIZATION

NRL Problem K03-08A
Project 625 03 01R, ARPA 660

Manuscript submitted October 23, 1967.

SECOND HARMONIC GENERATION BY REFLECTION FROM GaAs

1. THE GENERAL PHASE MATCHING PROBLEM

The onset of stimulated effects in nonlinear optics is quite dramatic. Ordinary nonlinear effects arising from even the most intense excitation are barely detectable whereas stimulated effects may be of the same order of intensity as the excitation. It may even be said that the extension of stimulated effects into the optical region is the cause of the recent explosion of interest in nonlinear optics. For the nonlinear effect, second harmonic generation, a criterion of analogous importance is that of phase match between the fundamental and second harmonic. When this condition is met, the two waves proceed in the same direction, in step, in principle allowing complete power transfer from the fundamental beam to the second harmonic beam. If this condition is not met, the second harmonic intensity remains very small even if a very intense fundamental is used.

Often a medium is normally dispersive and the index of refraction at the second harmonic is higher than at the fundamental. Then it is not possible for the two waves to proceed in the same direction and be in step for the magnitude of their wave vectors is different. The most well known way of achieving phase match is to use the birefringence of certain anisotropic, nonabsorbing, nonlinear crystals.[1] In an anisotropic medium there are in general two indices of refraction for each direction and they vary with direction. Thus sometimes a direction can be found where an index at the fundamental is the same as at the second harmonic. Recently the use of optical activity has been advanced as a means of achieving phase match.[2] Both of these methods make use of linear optical properties, which may be comprehensively categorized by the use of matrix algebra. If A' is the 2 x 2 matrix obtained by projecting A in $\underset{\sim}{E} = A\underset{\sim}{D}$ onto the plane transverse to the wave vector, then the 1) real symmetric, 2) real antisymmetric, 3) imaginary symmetric, 4) imaginary antisymmetric parts of A' contribute respectively the following optical effects: 1) birefringence, 2) optical activity, 3) anisotropic absorbtion, 4) circular dichoism.[3] In general optical media exhibit all combinations of these effects. From this it is clear that if the two methods above represent fairly the state of results in the general phase matching problem, as the author believes, then it is not completely solved.

This report is a calculation of the second harmonic intensity produced by reflection from a Gallium Arsenide crystal, including the effects of absorbtion at both fundamental and second harmonic.

[1] D. A. Kleinman, Phys. Rev. 128, 1761 (1962).
[2] H. Rabin and P. Bey, Bull. Am. Phys. Soc. 12, 81 (1967).
[3] G. Ramachandran and S. Ramaseshan, Handbuch der Physik XXV/1 107 (1961).

Conceived of as an approach to the general phase matching problem, second harmonic production by reflection is rather oblique. In fact it is not even a true subcase since the parametric approximation used here is not capable of eliciting the effects at phase match. Still it exhibits many necessary techniques and a thorough understanding of it is no doubt a prerequisite for an attack on the general phase matching problem. Thus while most of the theory appearing here has appeared before,[4] general methods are used where known because of the above outlook on the general phase matching problem.

The essential steps in the calculation of the reflected second harmonic intensity, each giving rise to a section below, are: describing GaAs and the geometry, solving the nonlinear wave equation, solving the boundary value problems and obtaining the intensities. The next to last section gives some numerical results and the last section remarks on the general phase matching problem.

[4] P. Butcher, Nonlinear Optical Phenomena, Ohio State University (1965)

2. GALLIUM ARSENIDE AND THE EXPERIMENTAL SETUP

An experiment with GaAs of the sort considered here was first reported in 1961.[5] Since GaAs is opaque, a reflection experiment appears to be required. It has $\bar{4}3m$ point group symmetry.[6] This point group is cubic so second order material tensors reduce to scalars and linear optical effects become isotropic.[7] Furthermore the point group excludes the antisymmetric part of a second order tensor so optical activity and circular dichroism are absent. Thus the linear optical properties of GaAs may be completely described by a complex dielectric susceptibility

$$\epsilon = (\nu + \iota \kappa)^2 \tag{2.1}$$

where ν and κ are the conventional index of refraction and absorbtion coefficient. Suppose the illumination is ruby laser light at 6943A. If the corresponding radian frequency is ω_o, then for present purposes the linear optical properties are completely described by[8]

[5] J. Ducuing and N. Bloembergen, Phys. Rev. Letters 10, 474 (1961).
[6] Wycoff Structure Tables Vol. I
[7] The latter part of this section is a discussion of the effects of symmetry on material tensors.
[8] J. Davey and T. Pankey, J. Appl. Phys. 35, 2203 (1964). The accuracy of these figures is probably a few percent of the largest figure. They were obtained by a Kramers-Kronig analysis of reflectance data of H. R. Philipp and H. Ehrenreich, Phys. Rev. 129, 1550 (1963).

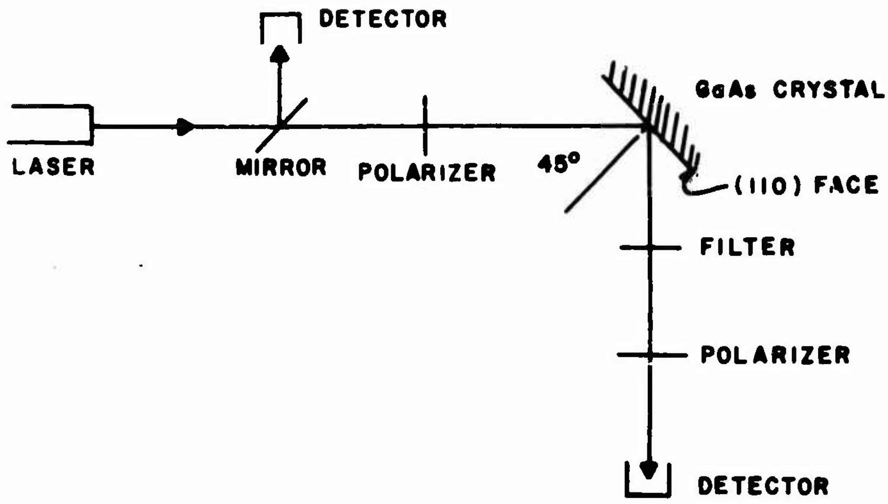

Fig. 1 - Experimental Set-up

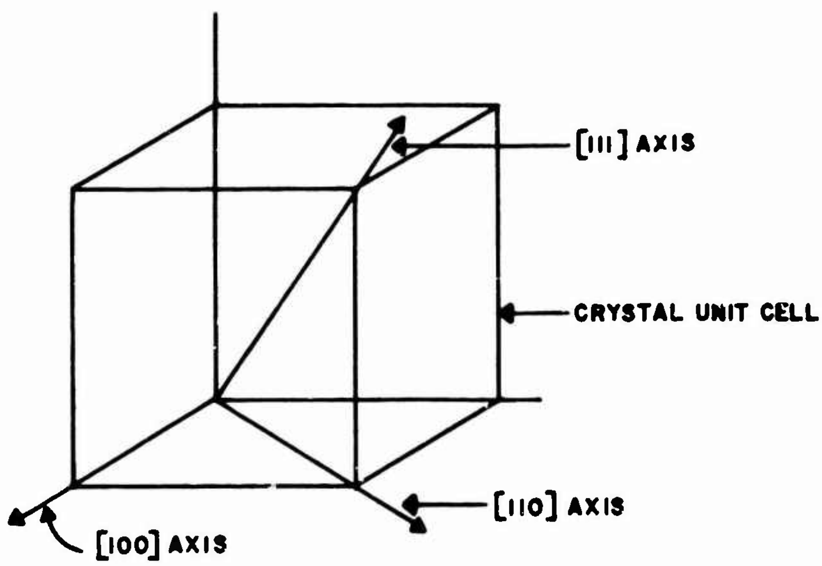

Fig. 2 - Crystal Axes

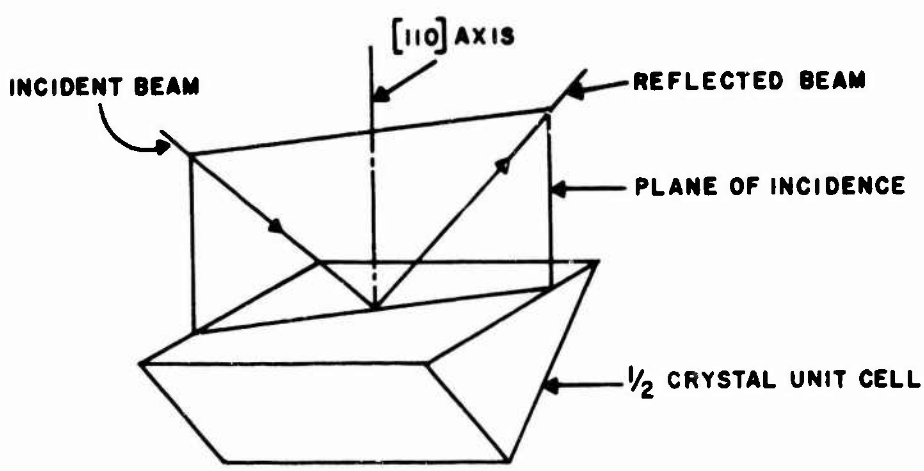

Fig. 3 - Crystal Orientation

$$\nu_1 = 3.79 \qquad \kappa_1 \cong .25$$
$$\nu_2 = 3.89 \qquad \kappa_2 = 1.53 \tag{2.2}$$

where ν_n is written for $\nu(n\omega_0)$.

Specifically the experiment considered is the following. A GaAs crystal is prepared with a (110) type plane as a reflecting surface in such a way as not to damage the crystalline properties of the surface layers. This is important since the absorption depths are

$$\lambda/\kappa_1 \cong 4400\text{A} \quad , \quad \lambda/\kappa_2 = 361\text{A}.$$

Ruby laser light of known intensity I and polarization, normal n or parallel p, strikes the reflecting surface with angle of incidence θ. The reflected beam is filtered to remove the fundamental. The second harmonic intensity is then measured in two polarization components Jn and Jp. The crystal may be rotated about a perpendicular to the surface to expose different crystal orientations. This orientation angle is Ψ and the $\Psi = 0$ reference will be specified later. These points are illustrated in Figs. 1-3.

From the foregoing and from general considerations one may write constitutive relations taylored to GaAs and the experiment. For a medium at optical frequencies

$$\underset{\sim}{B} = \underset{\sim}{H} \tag{2.3}$$

and assuming no space dispersion one has locally

$$D(t) = E(t) + \int_{-\infty}^{\infty} d\tau R^{(1)}(t - \tau)E(\tau)$$

$$+ \int_{-\infty}^{\infty} d\tau_1 \int_{-\infty}^{\infty} d\tau_2 R^{(2)}(t - \tau_1, t - \tau_2)E(\tau_1)E(\tau_2) + \ldots \tag{2.4}$$

where $R^{(1)}$ and $R^{(2)}$ vanish for negative arguments and thus give a casual relation. This is a relation between vector components with component indices temporarily omitted. Thus $R^{(1)}$ is a second order tensor and $R^{(2)}$, third. The use of the Taylor expansion excludes hard nonlinearities (multivalued regions, jumps, infinities) in order to avoid overly restrictive domains of convergence. Furthermore truncating (2.4) after the last term written is allowed in the small field approximation. In the frequency domain, this gives

$$D(\omega) = E(\omega) + \chi^{(1)}(\omega)E(\omega)$$

$$+ \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2 \, \delta(\omega - \omega_1 - \omega_2)\chi^{(2)}(\omega_1, \omega_2)E(\omega_1)E(\omega_2), \tag{2.5}$$

and $\chi^{(1)}$ and $\chi^{(2)}$ are analytic when considered as functions of complex arguments. In the time domain fields must be real and this imposes, in the frequency domain

$$E^*(\omega) = E(-\omega). \tag{2.6}$$

For E(t) and D(t) simultaneously real

$$\chi^{(1)*}(\omega) = \chi^{(1)}(-\omega), \tag{2.7}$$

$$\chi^{(2)*}(\omega_1, \omega_2) = \chi^{(2)}(-\omega_1, -\omega_2). \tag{2.8}$$

The discussion will now be restricted to the steady state case where frequencies present at one time are always present. Let the incident beam be of the form

$$E(t) = Ee^{-\iota\omega_0 t} + cc,$$

where cc represents the complex conjugate of the preceeding term. In the frequency domain,

$$E(\omega) = 2\pi\delta(\omega - \omega_0)E + 2\pi\delta(\omega + \omega_0)E^*$$

$$= 2\pi\delta(\omega - \omega_0)E + \widetilde{cc} \tag{2.9}$$

where $\widetilde{cc}$ implies that (2.9) satisfies (2.6). Substituting in (2.5)

$$D(\omega) = 2\pi\delta(\omega - \omega_o)[1 + \chi^{(1)}(\omega_o)]E + \tilde{cc}$$

$$+ 2\pi\delta(\omega - 2\omega_o)\chi^{(2)}(\omega_o, \omega_o)E^2 + \tilde{cc}$$

$$+ 2\pi\delta(\omega)\chi^{(2)}(\omega_o, -\omega_o)EE^* + \tilde{cc},$$

using (2.7) and (2.8). Subsequently the $\omega = 0$ component will be ignored. Reverting to the time domain, holding ω_o constant,

$$D(t) = \epsilon(\omega_o)Ee^{-\iota\omega_o t} + cc + 4\pi P_{NL}(2\omega_o, t) \tag{2.10}$$

where $$\epsilon(\omega_o) = 1 + \chi^{(1)}(\omega_o) \tag{2.11}$$

is the usual susceptibility and

$$P_{NLi}(2\omega_o, t) = \chi_{ijk}{}^{(2)}(\omega_o, \omega_o)E_jE_k e^{-2\iota\omega_o t} + cc \tag{2.12}$$

Summation on repeated indices is implied. Henceforth NL will be omitted since the linear polarization is not mentioned. Evidently the incident wave excites second harmonic polarization of the medium and, as will be seen in the next section, second harmonic radiation is emitted.

It has been mentioned that ϵ reduces to a scalar because of crystal symmetry. Likewise the second order susceptibility $\chi^{(2)}$, having at most 27 independent components, undergoes considerable reduction.[9] As an example of reduction due to symmetry, consider the symmetry

$$\chi_{ijk}^{(2)}(\omega_o, \omega_o) = \chi_{ikj}^{(2)}(\omega_o, \omega_o),$$

obtained from (2.12). The 9 elements of the form $\chi_{ijj}^{(2)}$ are unaffected but the remaining 18 are equal in pairs. Thus it should require at most 18 constants to specify $\chi_{ijk}^{(2)}$. Applying the elements of $\bar{4}3m$ to the remaining elements reduces them to 6 equal elements while the remainder vanish. There are several listings of tensors reduced by crystal symmetry in the literature. One such for third order tensors gives $S_{14} = S_{25} = S_{36}$.[10] In this "contracted" notation one writes the components symmetric in the last two indices separately from those antisymmetric. Under this convention the second index means 1:xx, 2:yy, 3:zz, 4:yz, 5:xz, 6:xy. Thus in present notation

$$\chi_{123} = \chi_{P(123)}$$

[9] A general treatment of this problem is P. Erdös, Helv. Phys. Acta 37, 493 (1964).
[10] J. Giordmaine, Phys. Rev. 138, A1601 (1965).

where $P(\cdot 123)$ is any permutation of 123. Henceforth χ represents χ_{123}. This reduction is performed in a special coordinate system in which a simple representation of the symmetry elements is possible. For many point groups, for cubic point groups in particular, the axes of this special system are the same as those of the conventional unit cell.[11] The result is for (2.12), writing the complex amplitudes of the positive frequency parts only

$$P_1 = \chi E_2 E_3,$$

$$P_2 = \chi E_1 E_3,$$

$$P_3 = \chi E_1 E_2,$$

a local relation in crystal coordinates.

This relation will now be evaluated in experimental coordinates by substituting components arising in two rotations. The rotation equations are written as substitutions, i.e., with old coordinates on the left,

$$\underset{\sim}{x} = R\underset{\sim}{x}'.$$

[11] International Tables for X-Ray Crystallography, Kynoch Press, Birmingham, England (1952) gives the convention for associating coordinate axes with conventional unit cells.

In this way one may drop the primes immediately as there is no further reference to the old coordinates. Let the first rotation be

$$R_1 = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & 1 \\ 0 & \sqrt{2} & 0 \\ -1 & 0 & 1 \end{pmatrix} = R_y\left(\frac{\pi}{4}\right)$$

since then the new Z axis is a [110] type axis. This gives

$$P_1 = -\chi E_2 E_1$$

$$P_2 = \frac{1}{2}\chi(E_3{}^2 - E_1{}^2)$$

$$P_3 = \chi E_2 E_3$$

Let the second rotation be

$$R_2 = \begin{pmatrix} \cos\Psi & -\sin\Psi & 0 \\ \sin\Psi & \cos\Psi & 0 \\ 0 & 0 & 1 \end{pmatrix} = R_z(\Psi)$$

to allow general rotations about the [110] axis. This gives

$$P_1 = -\chi[E_1 E_2(\cos^2\Psi - \sin^2\Psi) + (E_1{}^2 - E_2{}^2)\sin\Psi\cos\Psi]\cos\Psi$$

$$+ \frac{1}{2}\chi(E_3{}^2 - E_1{}^2\cos^2\Psi + 2E_1E_2\sin\Psi\cos\Psi - E_2{}^2\sin^2\Psi)\sin\Psi$$

$$P_2 = \chi[E_1E_2(\cos^2\Psi - \sin^2\Psi) + (E_1^2 - E_2^2)\sin\Psi\cos\Psi]\sin\Psi$$

$$+ \tfrac{1}{2}\chi(E_3^2 - E_1^2\cos^2\Psi + 2E_1E_2\sin\Psi\cos\Psi - E_2^2\sin^2\Psi)\cos\Psi$$

$$P_3 = \chi E_3(E_1\sin\Psi + E_2\cos\Psi).$$

The plane of incidence is the XZ plane and Ψ is the angle from the (100) plane to the plane of incidence. It is convenient to reduce this to cases of incident field polarization normal and parallel to the plane of incidence.

$$\left.\begin{aligned} P_1 &= \tfrac{1}{2}\chi E_2^2\sin\Psi(3\cos^2\Psi - 1) \\ P_2 &= -\tfrac{3}{2}\chi E_2^2\sin^2\Psi\cos\Psi \\ P_3 &= 0 \end{aligned}\right\} E_n(E_1 = 0 = E_3) \qquad (2.13)$$

$$\left.\begin{aligned} P_1 &= -\frac{3}{2}\chi E_1^2\sin\Psi\cos^2\Psi + \frac{1}{2}\chi E_3^2\sin\Psi \\ P_2 &= \tfrac{1}{2}\chi E_1^2\cos\Psi(3\sin^2\Psi - 1) + \tfrac{1}{2}\chi E_3^2\cos\Psi \\ P_3 &= \chi E_1E_3\sin\Psi \end{aligned}\right\} E_p(E_2 = 0) \qquad (2.14)$$

3. THE NONLINEAR WAVE EQUATION

The Maxwell equations for media at optical frequencies are

$$\nabla \times \underset{\sim}{H} = \frac{1}{c}\dot{\underset{\sim}{B}}, \qquad \nabla \cdot \underset{\sim}{B} = 0,$$
$$\nabla \times \underset{\sim}{E} = -\frac{1}{c}\dot{\underset{\sim}{D}}, \qquad \nabla \cdot \underset{\sim}{D} = 0. \tag{3.1}$$

Applying (2.3) to (3.1) and eliminating $\underset{\sim}{B}$ and $\underset{\sim}{H}$ gives the wave equation

$$\nabla \times \nabla \times \underset{\sim}{E} + \frac{1}{c^2}\ddot{\underset{\sim}{D}} = 0,$$

which gives in the frequency domain

$$\nabla \times \nabla \times \underset{\sim}{E}(\omega) - \frac{\omega^2}{c^2}\underset{\sim}{D}(\omega) = 0. \tag{3.2}$$

This equation is nonlinear since $\underset{\sim}{D}$ is nonlinear in $\underset{\sim}{E}$ so the method of eigenfunctions, in which the general solution is obtained by superposition, does not work. Thus $\underset{\sim}{E}(\omega)$ must be the total field. One anzatz for the form of the total field is

$$\underset{\sim}{E}(\omega) = 2\pi \sum_{n=-\infty}^{\infty} \delta(\omega - n\omega_o)\underset{\sim}{E}_n \tag{3.3}$$

because once one frequency is introduced the sum and difference effects of (2.5) may build up all harmonics. Introducing (3.3) in (3.2) gives an infinite system of connected differential equations for which the incident field $\underset{\sim}{E}_{\pm 1}$ is known. An iteration procedure may be set up based on

$$E_n = \sum_{r=a(n)}^{\infty} \lambda^r E_n^{(r)}$$

where r is the order of the iteration, λ is an expansion parameter to be set equal to one, and a(n) is the order of first appearance of the $n^{\underline{th}}$ harmonic. The zeroth iteration is

$$\nabla \times \nabla \times \underset{\sim}{E}_1^{(0)} - \frac{\omega_o^2}{c^2} \epsilon_1 \underset{\sim}{E}_1^{(0)} = 0 \qquad (3.4)$$

where $\epsilon_n = \epsilon(n\omega_o)$. The first iteration is

$$\nabla \times \nabla \times \underset{\sim}{E}_2^{(1)} - 4 \frac{\omega_o^2}{c^2} \epsilon_2 \underset{\sim}{E}_2^{(1)} = 16\pi \frac{\omega_o^2}{c^2} \underset{\sim}{P}(\underset{\sim}{E}_1^{(0)}) \qquad (3.5)$$

and there will be an $\omega = 0$ contribution in this order also. The next iteration gives a correction to the fundamental, $\underset{\sim}{E}_1^{(2)}$, and the third harmonic appears, $\underset{\sim}{E}_3^{(2)}$. A correction to the second harmonic does not appear until the third iteration and it is third order in $\chi^{(2)}$, a very small quantity. Thus

(3.5) gives an accurate $\underset{\sim}{E}_2$. This approximation is called the parametric approximation since $\underset{\sim}{E}_1$ evidently may be thought of as a source of energy which generates $\underset{\sim}{E}_2$ via the parameter $\chi^{(2)}$. If significant depletion of $\underset{\sim}{E}_1$ occurs, the approximation is invalid. Therefore phase matching for any harmonic actually present must be excluded. As a result the incident wave behaves as it would in a linear medium and the second harmonic is obtained as the solution of a linear inhomogeneous wave equation with an effectively known source term $\underset{\sim}{P}(\underset{\sim}{E}_1)$.

Using the complex amplitudes of the positive frequency parts of (2.12), the source term has the plane wave form

$$P_i e^{\iota \underset{\sim}{k}(\omega_0) \cdot \underset{\sim}{x}} = \chi_{ijk} E_{1j} e^{\iota \underset{\sim}{k}(\omega_0) \cdot \underset{\sim}{x}} E_{1k} e^{\iota \underset{\sim}{k}(\omega_0) \cdot \underset{\sim}{x}}$$

hence
$$\underset{\sim}{k}(2\omega_0) = 2\underset{\sim}{k}(\omega_0) \tag{3.6}$$

and
$$P_i = \chi_{ijk} E_{1j} E_{1k}$$

Because of homogeneity these amplitudes are constant throughout the medium. Variation due to absorption is accomplished by using a complex wave vector. For (3.5) then[12]

[12] Note that vector magnitudes are written $|\underset{\sim}{k}| = k$.

$$[(k_2^{\ 2} - 4\frac{\omega_o^{\ 2}}{c^2}\epsilon_2)I - \underset{\sim}{k}_2\underset{\sim}{k}_2] \cdot \underset{\sim}{E}_2 = 16\pi\frac{\omega_o^{\ 2}}{c^2}\underset{\sim}{P}$$

where I is the unit diadic. Since any vector may be decomposed into parts longitudinal and transverse to a direction $\hat{\underset{\sim}{k}}_2$,[13]

$$\underset{\sim}{P} = (I - \hat{\underset{\sim}{k}}_2\hat{\underset{\sim}{k}}_2) \cdot \underset{\sim}{P} + \hat{\underset{\sim}{k}}_2\hat{\underset{\sim}{k}}_2 \cdot \underset{\sim}{P}$$

$$= \hat{\underset{\sim}{k}}_2 \times (\underset{\sim}{P} \times \hat{\underset{\sim}{k}}_2) + \hat{\underset{\sim}{k}}_2\hat{\underset{\sim}{k}}_2 \cdot \underset{\sim}{P}.$$

Decomposing $\underset{\sim}{E}_2$ similarly, one may equate longitudinal and transverse parts.

$$-4\frac{\omega_o^{\ 2}}{c^2}\epsilon_2\hat{\underset{\sim}{k}}_2\hat{\underset{\sim}{k}}_2 \cdot \underset{\sim}{E}_2 = 16\pi\frac{\omega_o^{\ 2}}{c^2}\hat{\underset{\sim}{k}}_2\hat{\underset{\sim}{k}}_2 \cdot \underset{\sim}{P}$$

$$(k_2^{\ 2} - 4\frac{\omega_o^{\ 2}}{c^2}\epsilon_2)\hat{\underset{\sim}{k}}_2 \times (\underset{\sim}{E}_2 \times \hat{\underset{\sim}{k}}_2) = 16\pi\frac{\omega_o^{\ 2}}{c^2}\hat{\underset{\sim}{k}}_2 \times (\underset{\sim}{P} \times \hat{\underset{\sim}{k}}_2)$$

or

$$\hat{\underset{\sim}{k}}_2 \cdot \underset{\sim}{E}_2 = -\frac{4\pi}{\epsilon_2}\hat{\underset{\sim}{k}}_2 \cdot \underset{\sim}{P} \qquad (3.7)$$

$$\hat{\underset{\sim}{k}}_2 \times \underset{\sim}{E}_2 = \frac{16\pi\omega_o^{\ 2}}{c^2k_2^{\ 2} - 4\omega_o^{\ 2}\epsilon_2}\hat{\underset{\sim}{k}}_2 \times \underset{\sim}{P}$$

$$= \frac{4\pi}{\epsilon_1 - \epsilon_2}\hat{\underset{\sim}{k}}_2 \times \underset{\sim}{P}. \qquad (3.8)$$

[13] Circumflex denotes unit vector.

($\epsilon_1 \neq \epsilon_2$ since phase matching has been excluded.) In the last step (3.6) was used and (3.10) was anticipated. To complete the solution any solution of the homogeneous equation may be added. Homogeneous solutions are

$$\underset{\sim}{k}(\omega) \cdot \underset{\sim}{E}_H(\omega) = 0, \tag{3.9}$$

$$\underset{\sim}{k}^2(\omega) = \frac{\epsilon(\omega)\omega^2}{c^2} \quad . \tag{3.10}$$

4. THE BOUNDARY VALUE PROBLEMS

In the coordinates set up in section 2, let the boundary be the plane $\underset{\sim}{\hat{n}} \cdot \underset{\sim}{x} = 0$ with $\underset{\sim}{\hat{n}}$ pointing into the first medium which is transparent and nondispersive. The second medium is described by section 2. Given an incident uniform plane wave with linear polarization, find the reflected second harmonic intensity. There are three steps. Find: 1) the transmitted fundamental, 2) the second harmonic polarization (section 2), and 3) the reflected second harmonic. This section concerns the first and last steps.

Rays are designated in Table 1 and Fig. 4. The first and second symbols on each line pertain respectively to the fundamental and second harmonic. Angles are measured from the vertical in the respective medium. In the boundary value problem for the fundamental first the wave vectors will be determined before dealing with the amplitudes.

ω_o / $2\omega_o$	Ampli-tude	Wave Vector	Angle of Incidence
Incident or Source Wave	$\underset{\sim}{E}$ $\underset{\sim}{S}$	$\underset{\sim}{k}$ $\underset{\sim}{s}$	θ σ
Transmitted Wave	$\underset{\sim}{F}$ $\underset{\sim}{T}$	$\underset{\sim}{\ell}$ $\underset{\sim}{t}$	ϕ τ
Reflected Wave	$\underset{\sim}{G}$ $\underset{\sim}{R}$	$\underset{\sim}{m}$ $\underset{\sim}{r}$	γ ρ

Table 1 Ray Nomenclature

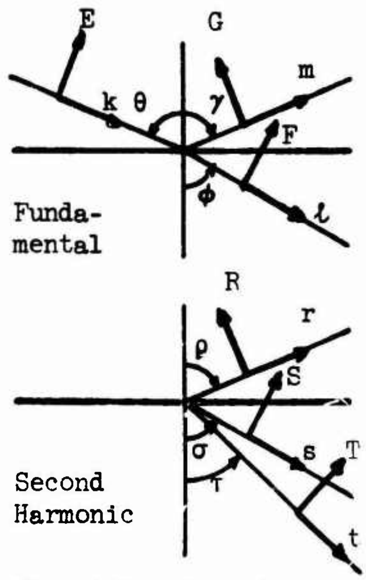

Fig. 4 Ray Geometry

From (3.10)

$$k = m = \frac{\omega_o}{c} \tag{4.1}$$

$$\frac{\epsilon_1 \omega_o^2}{c^2} = \ell^2 = \ell'^2 - \ell''^2 + 2i \underset{\sim}{\ell}' \cdot \underset{\sim}{\ell}'' \tag{4.2}$$

where $\ell = \ell' + i\ell''$, ℓ' and ℓ'' real. Since the waves must be the same function of position on the boundary,

$$\underset{\sim}{k} \cdot \underset{\sim}{x} = \underset{\sim}{\ell}' \cdot \underset{\sim}{x} = \underset{\sim}{m} \cdot \underset{\sim}{x}, \qquad \hat{\underset{\sim}{n}} \cdot \underset{\sim}{x} = 0 \tag{4.3}$$

and

$$0 = \underset{\sim}{\ell}'' \cdot \underset{\sim}{x}, \qquad \hat{\underset{\sim}{n}} \cdot \underset{\sim}{x} = 0. \tag{4.4}$$

The latter implies

$$\hat{\underset{\sim}{\ell}}'' = -\hat{\underset{\sim}{n}} \tag{4.5}$$

and

$$\underset{\sim}{\ell}' \cdot \underset{\sim}{\ell}'' = \ell'\ell''\cos\phi \tag{4.6}$$

since the transmitted wave is nonuniform due to absorption and decreases most rapidly in the direction $-\hat{\underset{\sim}{n}}$. Using the vector identity

$$\hat{\underset{\sim}{n}} \times (\underset{\sim}{x} \times \hat{\underset{\sim}{n}}) = \underset{\sim}{x} - \hat{\underset{\sim}{n}} \cdot \underset{\sim}{x}\hat{\underset{\sim}{n}}$$

or

$$\underset{\sim}{x} = \hat{\underset{\sim}{n}} \times (\underset{\sim}{x} \times \hat{\underset{\sim}{n}}), \qquad \hat{\underset{\sim}{n}} \cdot \underset{\sim}{x} = 0,$$

(4.3) becomes

$$\underset{\sim}{k} \times \hat{\underset{\sim}{n}} \cdot \underset{\sim}{x} \times \hat{\underset{\sim}{n}} = \underset{\sim}{\ell}' \times \hat{\underset{\sim}{n}} \cdot \underset{\sim}{x} \times \hat{\underset{\sim}{n}} = \underset{\sim}{m} \times \hat{\underset{\sim}{n}} \cdot \underset{\sim}{x} \times \hat{\underset{\sim}{n}}.$$

But since a vector of the form $\underset{\sim}{A} \times \hat{\underset{\sim}{n}}$ is in the surface and since $\underset{\sim}{x} \times \hat{\underset{\sim}{n}}$ is any surface vector,

$$\underset{\sim}{k} \times \underset{\sim}{\hat{n}} = \underset{\sim}{\ell}' \times \underset{\sim}{\hat{n}} = \underset{\sim}{m} \times \underset{\sim}{\hat{n}} \tag{4.7}$$

or

$$k\sin\theta = \ell'\sin\phi = m\sin\gamma \ . \tag{4.8}$$

Being real, $\underset{\sim}{m}$ is now completely determined by the magnitude (4.1), the two components (4.7) and by knowing in which medium it lies. Then

$$\gamma = \theta . \tag{4.9}$$

Also (4.7) states that $\underset{\sim}{k}$, $\underset{\sim}{\ell}'$, $\underset{\sim}{m}$, and $\underset{\sim}{\hat{n}}$ lie in a common plane, defined to be the plane of incidence. To complete the determination of $\underset{\sim}{\ell}$ define

$$n_1' = \frac{c\ell'}{\omega_o} , \qquad n_1'' = \frac{c\ell''}{\omega_o} . \tag{4.10}$$

Then (4.2) and (4.6) give

$$\epsilon_1 = n_1'^2 - n_1''^2 + 2in_1'n_1''\cos\phi$$

and using (4.8)

$$\cos = \frac{1}{n_1'}\sqrt{n_1'^2 - \sin^2\theta}, \qquad \sin\theta < n_1' ,$$

taking the positive square root since by (4.5), $\underset{\sim}{\ell}'$ and $\underset{\sim}{\ell}''$ lie in the same quadrant. This gives

$$\epsilon_1 = n_1'^2 - n_1''^2 + 2i n_1'' \sqrt{n_1'^2 - \sin^2\theta} \qquad (4.11)$$

together with the restriction

$$\sin\theta < n_1', \qquad (4.12)$$

excluding total reflection. The inverse of (4.11) is

$$\left.\begin{aligned} 2n_1'^2 &= \epsilon_1' + \sin^2\theta + \sqrt{(\epsilon_1' - \sin^2\theta)^2 + \epsilon_1''^2} \\ 2n_1''^2 &= -\epsilon_1' + \sin^2\theta + \sqrt{(\epsilon_1' - \sin^2\theta)^2 + \epsilon_1''^2} \end{aligned}\right\} \qquad (4.13)$$

giving the "refractive index" and "absorption coefficient" in terms of the complex susceptibility and angle of incidence. Since the definition (4.10) makes n_1' and n_1'' dependent on θ, they are not strictly material constants but depend on a non-material boundary condition as well. The usual convention is (2.1) which is used here only to determine ϵ from (2.2). Now $\underset{\sim}{\ell}'$ and $\underset{\sim}{\ell}''$ are determined by (4.10), (4.7), and (4.5) just as $\underset{\sim}{m}$ was. Because of medium homogeneity this determination of $\underset{\sim}{\ell}$ and $\underset{\sim}{m}$ applies throughout the appropriate medium.

The amplitudes may be determined by the tangential boundary conditions for electric and magnetic vectors,

$$(\underset{\sim}{E} + \underset{\sim}{G} - \underset{\sim}{F}) \times \hat{\underset{\sim}{n}} = 0,$$

$$(\underset{\sim}{k} \times \underset{\sim}{E} + \underset{\sim}{m} \times \underset{\sim}{G} - \underset{\sim}{\ell} \times \underset{\sim}{F}) \times \hat{\underset{\sim}{n}} = 0,$$

since according to (3.9) they are transverse,

$$\underset{\sim}{k} \cdot \underset{\sim}{E} = \underset{\sim}{m} \cdot \underset{\sim}{G} = \underset{\sim}{\ell} \cdot \underset{\sim}{F} = 0.$$

Using the experimental coordinates of section 2, one has

$$k_2 = m_2 = \ell_2 = 0$$

and a system of 6 complex algebraic equations in 6 complex unknowns, $\underset{\sim}{F}$ and $\underset{\sim}{G}$.

$$\left.\begin{aligned}
F_1 - G_1 &= E_1 \\
F_2 - G_2 &= E_2 \\
\ell_3 F_2 - m_3 G_2 &= k_3 E_2 \\
\ell_3 F_1 - \ell_1 F_3 - m_3 G_1 + m_1 G_3 &= k_3 E_1 - k_1 E_3 \\
\ell_1 F_1 + \ell_3 F_3 &= 0 \\
m_1 G_1 + m_3 G_3 &= 0
\end{aligned}\right\} \qquad (4.14)$$

Before solving by Cramer's rule it is necessary to see if the system determinant has any significant zeroes.

$$0 = \det \begin{vmatrix} 1 & 0 & 0 & -1 & 0 & 0 \\ 0 & 1 & 0 & 0 & -1 & 0 \\ 0 & \ell_3 & 0 & 0 & -m_3 & 0 \\ \ell_3 & 0 & -\ell_1 & -m_3 & 0 & m_1 \\ \ell_1 & 0 & \ell_3 & 0 & 0 & 0 \\ 0 & 0 & 0 & m_1 & 0 & m_3 \end{vmatrix} = \det \begin{vmatrix} 1 & -1 & 0 & 0 & 0 & 0 \\ \ell_3 & -m_3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & -1 & 0 \\ 0 & 0 & \ell_3 & -\ell_1 & -m_3 & m_1 \\ 0 & 0 & \ell_1 & \ell_3 & 0 & 0 \\ 0 & 0 & 0 & 0 & m_1 & m_3 \end{vmatrix}$$

Thus the system is block diagonal and separates into two independent systems, one for y field components only and one for x and z components. For zeroes of the y system determinant

$$0 = \det \begin{vmatrix} 1 & -1 \\ \ell_3 & -m_3 \end{vmatrix} = \ell_3 - m_3$$

or

$$\ell_3' = m_3 \text{ and } \ell_3'' = 0 .$$

Using (4.5) the latter implies $\underset{\sim}{\ell}'' = 0$ so from (4.10) either $\omega_0 = 0$ or $n_1'' = 0$. In addition

$$\ell_3' = -\ell' \cos\phi$$

$$m_3 = m\cos\gamma$$

so from (4.1), (4.2), (4.9), (4.10) and $\cos\theta = -\cos\gamma$

$$n_1'\cos\phi = \cos\theta, \qquad \omega_0 \neq 0$$

$$= n_1' \sqrt{1 - \sin^2\phi}$$

$$= \sqrt{n_1'^2 - \sin^2\theta}$$

which implies either $\omega_0 = 0$ or $n_1' = 1$. Neither of these conditions, $\omega_0 = 0$ or $n_1' = 1$ and $n_1'' = 0$, is of present interest.

For the x and z system determinant to vanish

$$0 = \det \begin{vmatrix} 1 & 0 & -1 & 0 \\ \ell_3 & -\ell_1 & -m_3 & m_1 \\ \ell_1 & \ell_3 & 0 & 0 \\ 0 & 0 & m_1 & m_3 \end{vmatrix} = \ell_3 m^2 - m_3 \ell^2 \qquad (4.15)$$

which implies either $\ell_3 = 0 = m_3$ or $m^2/m_3 = \ell^2/\ell_3$ and the former is of no interest. Since both sides of the latter may be varied independently, its general solution is that both parts are equal to a common constant, say K. For the left hand side then K is real and

$$m^2 = m_1^2 + m_3^2 = Km_3$$

or

$$m_1^2 + (m_3 - \tfrac{1}{2}K)^2 = \tfrac{1}{4}K^2,$$

thus possible m_1 are $|m_1| \leq \frac{1}{2}K$. For the right hand side

$$\ell^2 = \ell_1^2 + \ell_3^2 = K\ell_3$$

and ℓ_3 is complex so

$$\ell_1^2 + \ell_3'^2 - \ell_3''^2 = K\ell_3', \qquad (4.16)$$

$$2\ell_3'\ell_3'' = K\ell_3''.$$

If $\ell_3'' \neq 0$ then $\ell_3' = \frac{1}{2}K$ and $\ell_1^{\,2} - \ell_3''^2 = \frac{1}{4}K^2$ so possible ℓ_1 are $|\ell_1| \geq \frac{1}{2}K$. From (4.16) this inequality holds even if $\ell_3'' = 0$. The only solution compatible with both inequalities and (4.8) is

$$|m_1| = \tfrac{1}{2}K = |\ell_1|$$

With (4.16) this implies $\ell_3'' = 0$, the no absorption case. Then from (4.1), (4.2), (4.15), and (4.8)

$$\ell^2 = \epsilon_1 m^2,$$

$$\ell_3 = \epsilon_1 m_3$$

$$\ell_1 = m_1$$

and eliminating ℓ's in the first equation

$$m_1^{\,2} + \epsilon_1^{\,2} m_3^{\,2} = \epsilon_1 (m_1^{\,2} + m_3^{\,2})$$

or

$$(\epsilon_1 - 1) m_1^{\,2} + (\epsilon_1 - \epsilon_1^{\,2}) m_3^{\,2} = 0$$

implying $\epsilon_1 = 1$ or $m_1^2 = \epsilon_1 m_3^2$ or

$$\tan^2\theta_B = \epsilon_1, \qquad \epsilon_1'' = 0. \tag{4.17}$$

This is the condition for Brewster's angle. For polarization in the plane of incidence, the reflected ray vanishes at this angle. It has been shown that this is the only significant zero of the system determinant.

Perhaps this investigation of null solutions is only a nicety in the present context. It does endow the calculation with a satisfactory feeling of completeness. However in more complicated cases this tool will still apply possibly to uncover regions of significant departure from the general solution, --regions which may be as useful as the Brewster's angle extinction.

To proceed with the general solution, the y component system gives by Cramer's rule

$$F_2 = \frac{k_3 - m_3}{\ell_3 - m_3} E_2 . \tag{4.18}$$

Likewise for the x component

$$F_1 = \frac{\frac{k^2}{k_3} - \frac{m^2}{m_3}}{\frac{\ell^2}{\ell_3} - \frac{m^2}{m_3}} E_1, \tag{4.19}$$

and from the transversality condition

$$F_3 = - \frac{\ell_1}{\ell_3} F_1 . \tag{4.20}$$

This completes the determination of the transmitted fundamental.

In the second harmonic boundary value problem the source wave vector is given by (3.6) and is complex.

$$\underset{\sim}{s} = 2\underset{\sim}{\ell} \tag{4.21}$$

The freely propagating wave vector magnitudes are from (3.10)

$$r = \frac{2\omega_0}{c} , \tag{4.22}$$

$$t^2 = \frac{4\epsilon_2 \omega_0^2}{c^2} . \tag{4.23}$$

Continuity of fields at the boundary requires equality of phase functions, giving for the complex parts

$$\underset{\sim}{s}'' \cdot \underset{\sim}{x} = \underset{\sim}{t}'' \cdot \underset{\sim}{x}, \qquad \underset{\sim}{\hat{n}} \cdot \underset{\sim}{x} = 0.$$

But by (4.4) and (4.21) $\underset{\sim}{s}''$ has no tangential part, hence

$$\underset{\sim}{\hat{t}}'' = - \underset{\sim}{\hat{n}}.$$

As before all wave vectors are in the plane of incidence and the real tangential components are determined by

$$t' \sin\tau = r \sin\rho = s' \sin\sigma. \tag{4.24}$$

The reflected wave vector is now completely determined and in particular from (4.21)

$$\sigma = \phi$$

and by comparing (4.24) and (4.8) using (4.22), (4.21), (4.2) and (4.1)

$$\rho = \theta. \tag{4.25}$$

Now define

$$n_2' = \frac{ct'}{2\omega_0}, \qquad n_2'' = \frac{ct''}{2\omega_0} \tag{4.26}$$

so that

$$\epsilon_2 = n_2'^2 - n_2''^2 + 2\iota n_2'' \sqrt{n_2'^2 - \sin^2\theta}$$

and total reflection does not occur. Inversely

$$\left.\begin{aligned} 2n_2'^2 &= \epsilon_2'^2 + \sin^2\theta + \sqrt{(\epsilon_2' - \sin^2\theta)^2 + \epsilon_2''^2} \\ 2n_2''^2 &= -\epsilon_2' + \sin^2\theta + \sqrt{(\epsilon_2' - \sin^2\theta)^2 + \epsilon_2''^2} \end{aligned}\right\} \quad (4.27)$$

Thus $\underset{\sim}{t}$ is also completely determined.

For the source wave amplitude one has from (3.7) and (3.8)

$$\underset{\sim}{\hat{s}} \cdot \underset{\sim}{S} = -\frac{4\pi}{\epsilon_2} \underset{\sim}{\hat{s}} \cdot \underset{\sim}{P} \quad (4.28)$$

$$\underset{\sim}{\hat{s}} \times \underset{\sim}{S} = \frac{4\pi}{\epsilon_1 - \epsilon_2} \underset{\sim}{\hat{s}} \times \underset{\sim}{P} \quad (4.29)$$

where $\underset{\sim}{P}$ is determined from $\underset{\sim}{F}$ by (2.13) and (2.14). Tangential boundary conditions are

$$(\underset{\sim}{S} + \underset{\sim}{T} - \underset{\sim}{R}) \times \underset{\sim}{\hat{n}} = 0$$

$$(\underset{\sim}{s} \times \underset{\sim}{S} + \underset{\sim}{t} \times T - \underset{\sim}{r} \times R) \times \underset{\sim}{\hat{n}} = 0$$

and from (3.11)

$$\underset{\sim}{t} \cdot \underset{\sim}{T} = \underset{\sim}{r} \cdot \underset{\sim}{R} = 0.$$

Except for the longitudinal component in the inhomogeneous term, (4.28), this system has the same algebraic form as before. For the y component, using (4.29) and $s_2 = 0$,

$$R_2 = \frac{\varepsilon_3 - t_3}{r_3 - t_3} S_2$$

$$= \frac{4\pi}{\epsilon_1 - \epsilon_2} \frac{s_3 - t_3}{r_3 - t_3} P_2 . \qquad (4.30)$$

For the x and z components

$$R_1 = \frac{r_3 (t^2 S_1 - t_3 S_{t2})}{t_3 r^2 - r_3 t^2}$$

$$R_3 = - \frac{r_1}{r_3} R_1 \qquad (4.31)$$

where

$$S_{t2} = s_3 S_1 - s_1 S_3$$

$$= \frac{4\pi}{\epsilon_1 - \epsilon_2} (s_3 P_1 - s_1 P_3),$$

the y component of (4.29). Solving (4.28) and (4.29) for S_1,

$$S_1 = -\frac{s_1}{s^2}\,\frac{4\pi}{\epsilon_2}\,(s_1P_1 + s_3P_3) + \frac{s_3}{s^2}\,\frac{4\pi}{\epsilon_1 - \epsilon_2}\,(s_3P_1 - s_1P_3)$$

$$= \frac{4\pi}{s^2\epsilon_2(\epsilon_1 - \epsilon_2)}\,[(\epsilon_2 s^2 - \epsilon_1 s_1{}^2)P_1 - \epsilon_1 s_1 s_3 P_3]$$

and using (4.21), (4.2), (4.23), and (4.24)

$$S_1 = \frac{4\pi\epsilon_1}{s^2\epsilon_2(\epsilon_1 - \epsilon_2)}\,[(t^2 - s_1{}^2)P_1 - s_1 s_3 P_3]$$

$$= \frac{4\pi}{t^2(\epsilon_1 - \epsilon_2)}\,[t_3{}^2P_1 - s_1 s_3 P_3].$$

Finally

$$R_1 = \frac{4\pi}{\epsilon_1 - \epsilon_2}\left(1 - \frac{s_3}{t_3}\right)\left[\frac{r^2}{r_3} - \frac{t^2}{t_3}\right]^{-1}\,[t_3P_1 + t_1P_3]. \qquad (4.32)$$

5. THE REFLECTED SECOND HARMONIC INTENSITY

A completely polarized beam can be analyzed by finding the intensities of two linearly polarized components and their phase difference. Arbitrarily limiting this calculation to an examination of intensities leaves as the most general set of measurements the four polarization cases listed in section 2. Furthermore all four cases provide different information from a qualitative standpoint. Thus in the last section the independence of the y component and x - z component systems, shows that in penetrating the surface, a ray preserves two special cases of linear polarization, normal or parallel to the plane of incidence. However the second harmonic polarization process mixes these components as shown by (2.13) and (2.14).

The normal component second harmonic intensity is , from (4.30)

$$J_n = \left| \frac{4\pi}{\epsilon_1 - \epsilon_2} \right|^2 \left| \frac{s_3 - t_3}{r_3 - t_3} \right|^2 |P_2|^2$$

with P_2 generated by $\underset{\sim}{F}$ through either (2.13) or (2.14) according to the incident polarization. Thus

$$J_{nn} = 9 \left| \frac{2\pi}{\epsilon_1 - \epsilon_2} \right|^2 \left| \frac{s_3 - t_3}{r_3 - t_3} \right|^2 |\chi|^2 \, |F_2|^4 \sin^4\Psi\cos^2\Psi$$

$$= 9 \left| \frac{2\pi}{\epsilon_1 - \epsilon_2} \right|^2 \left| \frac{s_3 - t_3}{r_3 - t_3} \right|^2 \left| \frac{k_3 - m_3}{\ell_3 - m_3} \right|^4 |\chi|^2 \, I_n^2 \sin^4\Psi\cos^2\Psi \quad (5.1)$$

where the second J index denotes incident polarization. Now

$$I_p = |E_1|^2 = |E_3|^2$$

so by the tranversality of $\underset{\sim}{E}$

$$|E_1|^2 = \frac{k_3^{\,2}}{k^2} I_p .$$

Using also (2.14), (4.19), (4.1) and (4.2)

$$|P_2|^2 = \tfrac{1}{4} |\chi|^2 \; |F_1^{\,2}\cos\Psi(3\sin^2\Psi - 1) + F_3^{\,2}\cos\Psi|^2$$

$$= \tfrac{1}{4} |\chi|^2 \left| \frac{m_3 k^2 - k_3 m^2}{m_3 \ell^2 - \ell_3 m^2} \right| |G_1|^2 \; I_p^{\,2}\cos^2\Psi |\; 1 + \frac{\ell_3^{\,2}}{\ell^2} (3\sin^2\Psi - 2)|^2,$$

hence

$$J_{np} = \left| \frac{2\pi\epsilon_1}{\epsilon_1 - \epsilon_2} \right|^2 \left| \frac{s_3 - t_3}{r_3 - t_3} \right|^2 \left| \frac{m_3 k^2 - k_3 m^2}{m_3 \ell^2 - \ell_3 m^2} \right|^4 |\chi|^2 I_p^{\,2}$$

$$\times \; \left|1 + \frac{\ell_3^{\,2}}{\ell^2} (3\sin^2\Psi - 2) \right|^2 \cos^2\Psi \qquad (5.2)$$

From (4.31) and (4.32)

$$J_p = |R_1|^2 = |R_3|^2$$

$$= \left|\frac{4\pi}{\epsilon_1 - \epsilon_2}\right|^2 \left|\frac{t_3 - s_3}{t_3 r^2 - r_3 t^2}\right|^2 |t_3 P_1 + t_1 P_3|^2 .$$

The case of normal component illumination simplifies because of the absence of a z component in (2.13).

$$J_{pn} = \left|\frac{2\pi}{\epsilon_1 - \epsilon_2}\right|^2 \left|\frac{t_3 - s_3}{t_3 r^2 - r_3 t^2}\right|^2 |t_s \chi F_2{}^2|^2 \sin^2\Psi (3\cos^2\Psi - 1)^2$$

$$= \left|\frac{2\pi}{\epsilon_1 - \epsilon_2}\right|^2 \left|\frac{t_3 - s_3}{t_3 r^2 - r_3 t^2}\right|^2 |t_3|^2 \left|\frac{k_3 - m_3}{\ell_3 - m_3}\right|^2 |\chi|^2 \times$$

$$\times \; I_n{}^2 \sin^2\Psi (3\cos^2\Psi - 1)^2 \qquad (5.3)$$

For parallel component illumination, from (2.14)

$$P_1 = \tfrac{1}{2}\chi(-3F_1{}^2\sin\Psi\cos^2\Psi + F_3{}^2\sin\Psi)$$

$$= \tfrac{1}{2}\chi\,\frac{\ell^2}{k_3{}^2}\left(\frac{m_3k^2 - k_3m^2}{m_3\ell^2 - \ell_3m^2}\right)^2 E_1{}^2\left[1 - \frac{\ell_3{}^2}{\ell^2}(3\cos^2\Psi + 1)\right]\sin\Psi$$

$$P_3 = \chi F_1F_3\sin\Psi$$

$$= -\chi\left(\frac{m_3k^2 - k_3m^2}{m_3\ell^2 - \ell_3m^2}\right)E_1{}^2\,\frac{\ell_1\ell_3}{k_3{}^2}\sin\Psi$$

hence

$$J_{pp} = \left|\frac{2\pi\epsilon_1}{\epsilon_1 - \epsilon_2}\right|^2\left|\frac{t_3 - s_3}{t_3r^2 - r_3t^2}\right|\left|\frac{m_3k^2 - k_3m^2}{m_3\ell^2 - \ell_3m^2}\right|^4 |\chi|^2I_p{}^2\sin^2\Psi$$

$$\cdot\left|t_3\left[1 - \frac{\ell_3{}^2}{\ell^2}(3\cos^2\Psi + 1)\right] - 2t_1\frac{\ell_1\ell_3}{\ell^2}\right|^2 \qquad (5.4)$$

6. NUMERICAL RESULTS

The above intensities are functions of a number of parameters,

$$J = J_{jk}(\omega_0, \epsilon_1, \epsilon_2, |\chi|, I_k, \theta, \Psi).$$

Also several factors are complex algebraic so one may feel it is hard to extract information at a glance. For discussion purposes define[14]

$$f(\theta, \Psi) = \left|1 - \frac{\ell_3^2}{\ell^2}(3\cos^2\Psi + 1) - 2\frac{\tau_1}{\tau_3}\frac{\ell_1\ell_3}{\ell^2}\right|^2 \sin^2\Psi \propto J_{pp}(\Psi) \quad (6.1)$$

$$g(\theta, \Psi) = \left|1 + \frac{\ell_3^2}{\ell^2}(3\sin^2\Psi - 2)\right|^2 \cos^2\Psi \propto J_{np}(\Psi) \quad (6.2)$$

These two factors contain all the Ψ dependence of two of the intensities and

$$f(0, \tfrac{\pi}{2} - \Psi) \propto J_{nn}(\Psi) \quad (6.3)$$

$$g(0, \tfrac{\pi}{2} - \Psi) \propto J_{pn}(\Psi) \quad (6.4)$$

contain the Ψ dependence of the other two. By changing the definition of Ψ from a geometrical one to a physical one it is possible to ascribe major features of the variation of intensity

[14] The symbol $\propto$ indicates proportionality.

with crystal orientation to crystalline features. Let Ψ' be the angle from [100] to the projection of the incident electric vector on the reflecting surface. Then in (6.3) and (6.4) $\Psi' = \frac{\pi}{2} - \Psi$ since the incident polarization is normal but in (6.1) and (6.2) $\Psi' = \Psi$. The curve $f(0, \Psi')$ shows extinctions on the [100] and [110] axes and a maximum on the [111] axis ($\Psi' = \cos^{-1} 3^{-\frac{1}{2}} = 54.7^\circ$). The curve $g(0, \Psi')$ shows extinctions at the [100] and [111] axes and an absolute maximum at the [110] axis. These two curves (see Fig. 5) are independent of θ and the material constants.

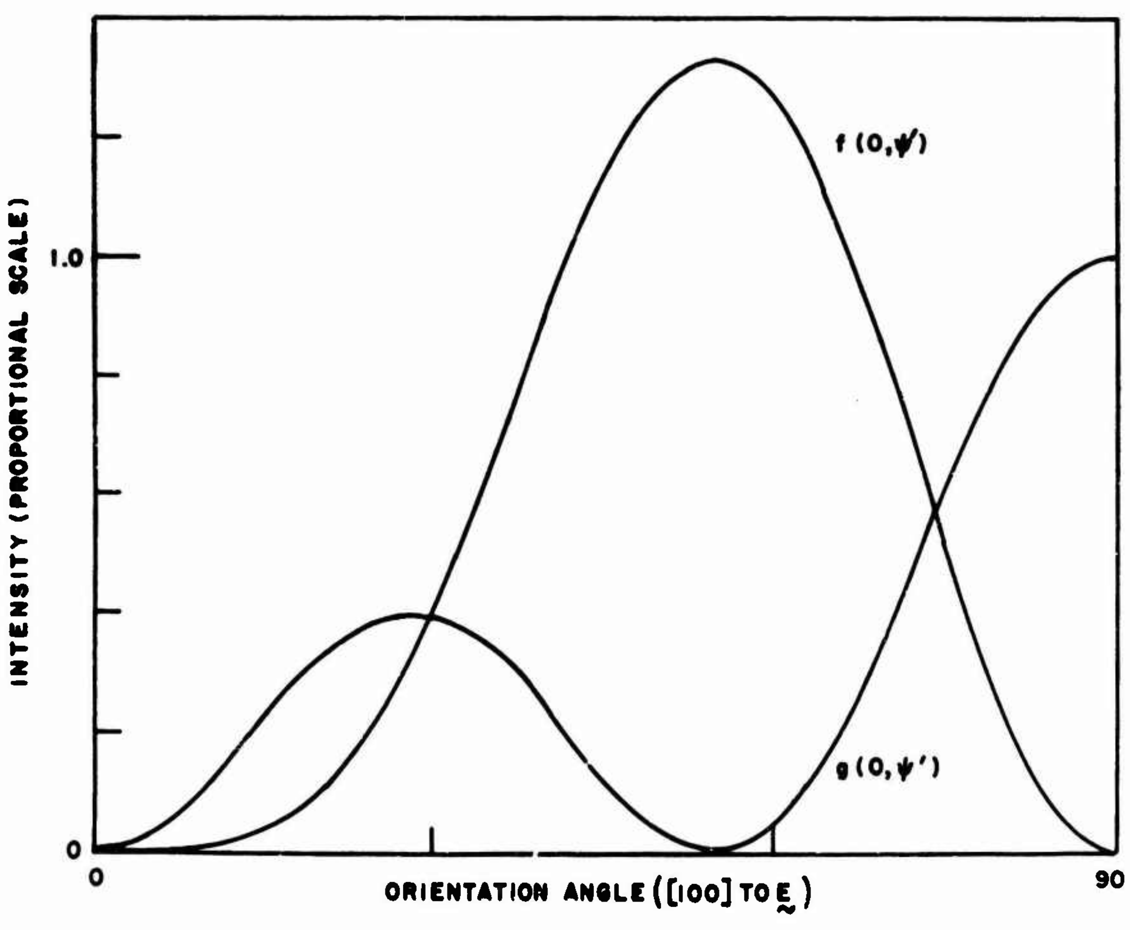

Fig. 5 - Variation of Intensity with Orientation

Using the frequency and material constants given in section 2, $f(\theta, \Psi)$ and $g(\theta, \Psi)$ have been computed for a representative sampling of points (θ, Ψ). The comparison of $f(0, \Psi)$ and $f(\theta, \Psi)$ may be regarded as exhibiting the difference in shape between $J_{nn}(\Psi')$ and $J_{pp}(\Psi)$. Likewise comparison of $g(0, \Psi)$ to $g(\theta, \Psi)$ exhibits the difference between $J_{pn}(\Psi')$ and $J_{np}(\Psi)$. Since the Ψ dependence is arbitrary to within a constant multiple, the comparison in shape is made by normalizing the maxima. For $\theta = 45^\circ$ the maximum variation in shape between $J_{nn}(\Psi')$ and $J_{pp}(\Psi)$ is 60% based on $f(0, \Psi)$ and the maximum variation in shape between $J_{pn}(\Psi')$ and $J_{np}(\Psi)$ is 14%. In both cases the maximum variation occurs when the projection of the incident electric vector falls between the [100] and [111] axes. For larger θ the differences increase. Thus though $J_{pp}(\Psi)$ and $J_{np}(\Psi)$ look more complicated, they have much the same shape as $J_{nn}(\Psi')$ and $J_{pn}(\Psi')$ respectively. As to specific details of difference $J_{pp}(\Psi)$ does not show a perfect extinction at [110] and neither does $J_{np}(\Psi)$ at [111]. Furthermore the maximum of $J_{pp}(\Psi)$ and the relative minimum of $J_{np}(\Psi)$ show a small shift from [111].

In addition f and g were computed in the approximations $\kappa_1 \cong 0$ and $\kappa_1 \cong 0 \cong \kappa_2$ which are convenient since they allow partial or complete shift to real arithmetic. At first glance the last approximation seems poor since it means neglecting $\kappa_2 = 1.53$ by comparison with $\nu_2 = 3.89$. The first approximation changes $f(45^\circ, \Psi)$ by .05% near its maximum and the second by 1.01% at the same point. In these approximations the largest changes relative to the exact values at the same points are 10.3% at $\Psi = 90^\circ$ and 24% near $\Psi = 85^\circ$. Both approximations are the same for $g(45^\circ, \Psi)$ and change it by .2%. It seems clear that once a very small absorption is admitted, increasing the absorption causes no qualitative change until the critical point $n'' = n'$ is reached, at which point propagation ceases. The latter is vaguely indicated above by failure of the analysis. Equation (4.13) requires $n_1' > n_1''$ unless $\epsilon_1' \leq 0$ and the latter causes a fundamental change in the character of the Helmhotz equation (3.4). Furthermore, even the transition from no absorption to some absorption appears smooth. The only qualitative change indicated above is the disappearance of the system null solution (4.17). Evidently when there is some absorption, the Brewster extinction becomes only a relative minimum.

The next most complicated factors in the intensities are recognizable as transmission ratios--one for transmission of the fundamental into the medium and one for transmission of the second harmonic out. The exact evaluation of the intensity coefficients for $\theta = 45^\circ$ gives the following.

$$J_{nn} = 2.926 \times 10^{-4}|\chi|^2 I_n{}^2 f(0, \Psi') \tag{6.5}$$

$$J_{np} = 6.279 \times 10^{-4}|\chi|^2 I_p{}^2 g(45^\circ, \Psi') \tag{6.6}$$

$$J_{pn} = 4.348 \times 10^{-4}|\chi|^2 I_n{}^2 g(0, \Psi') \tag{6.7}$$

$$J_{pp} = 9.329 \times 10^{-4}|\chi|^2 I_p{}^2 f(45^\circ, \Psi') \tag{6.8}$$

The following table lists per cent errors in the numerical coefficients in the approximations indicated.

	J_{nn}	J_{np}	J_{pn}	J_{pp}
$\chi_1 \cong 0$	2.2	2.0	2.1	2.0
$\kappa_1 \cong 0 \cong \kappa_2$	19.6	6.7	14.4	14.5

Table 2. Errors in Two Approximations of the Intensities

The conclusions of the last paragraph on the general effects of absorption still apply. Thus absorption considerations can be neglected if only qualitative behavior is sought.

Much of the behavior expected has been verified by experiment. The variation of J_{nn} and J_{pn} with Ψ on an arbitrary intensity scale were reported in 1961 by Ducuing and Bloembergen.[15] Certain aspects of the variation in θ have been obtained including the minimum near the Brewster extinction.[16] Finally $|\chi|$ relative to that of potassium dihydrogen phosphate has been obtained at several frequencies and for other semiconductors and the phase of χ for GaAs has been obtained at ruby laser frequency.[17] The data on $|\chi|$ is sufficient to show its general dispersion characteristics and it is seen that peaks of $\chi^{NL}(2\omega)$ roughly match those in $\chi^{L}(\omega)$.

[15] See footnote [5]. The same results appear in N. Bloembergen, Nonlinear Optics, Benjamin 1965, pp. 126-7 with better identification of polarization cases.
[16] R. K. Chang and N. Bloembergen, Phys. Rev. 144, 775 (1966)
[17] R. K. Chang, J. Ducuing and N. Bloembergen, Phys. Rev. Letters 15, 415 (1965). For a review of reflected second harmonic experiments, see N. Bloembergen, Optica Acta 13, 311 (1966).

7. REMARKS ON THE GENERAL PHASE MATCHING PROBLEM

In the first section the possibility that the output signal might attain intensities of the same order of magnitude as the pump signal was mentioned. When the output signal is the second harmonic, phase match is known to be a necessary condition for this possibility.[18] In this exposition of second harmonic generation by reflection, phase match occurred as a null solution to the nonlinear wave equation, i.e., $\epsilon_1 = \epsilon_2$ in (3.8). Because the concept of null solution is quite general, there is a strong inference that it is an avenue for extending those implications of phase matching which are contained in the above necessary condition. Thus it should be possible to generalize the results listed on pp. 1-2 for anisotropic nonabsorbing media and for optically active nonabsorbing media to anisotropic media exhibiting the most general linear optical effects.

The character of this generalization may be sketched in outline. It will be matrix algebraic and so will not differ much from Kleinman's approach.[1] If the indications of the last section on the relative unimportance of absorptive effects hold, the generalization of the work on birefringent media to include anisotropic absorption or the generalization of the work on optical activity to include circular dichroism is trivial. However, as a survey of effects attributable to the imaginary part of the dielectric

[18] J. A. Armstrong, N. Bloembergen, J. Ducuing and P. S. Pershan Phys. Rev. 127, 1930 (1962).

constant, the present effort is incomplete due partly to the exclusion of total reflection and partly to the lack of special consideration of the relation between optical activity and circular dichroism. Thus it will be necessary to complete the description of the dielectric constant. These generalizations still leave two classes of optical effects disjoint under the capability of describing phase match in terms of material properties. The two classes could be called symmetric and antisymmetric.[19] Since the dielectric constant for optical activity requires wave vector dependence, perhaps these two classes can be merged by generalizing the anzatz (2.4) to include spatial dispersion.

ACKNOWLEDGMENTS

The author wishes to thank Mr. S. Podgor for computer work and Dr. A. W. Sáenz for many helpful discussions.

[19] See the categorization of linear optics, p. 2.

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) Naval Research Laboratory Washington, D.C. 20390	2a. REPORT SECURITY CLASSIFICATION Unclassified 2b. GROUP

3. REPORT TITLE

SECOND HARMONIC GENERATION BY REFLECTION FROM GaAs

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

An interim report on one phase of the problem.

5. AUTHOR(S) (First name, middle initial, last name)

James B. Langworthy

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
January 19, 1968	49	18

8a. CONTRACT OR GRANT NO. NRL Problem K03-08A b. PROJECT NO. 625 03 01R, ARPA 660 c. d.	9a. ORIGINATOR'S REPORT NUMBER(S) NRL Report 6662 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)

10. DISTRIBUTION STATEMENT

This document has been approved for public release and sale; its distribution is unlimited.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY Dept. of Defense (Advanced Research Projects Agency) Washington, D.C. 20301

13. ABSTRACT

The intensity of second harmonic light generated by reflection from a Gallium Arsenide Crystal is calculated in detail in four cases of polarization. Relevant properties of GaAs are stated and an experiment is described. Based on this, constitutive relations are written in the weak field approximation and the second and third order material tensors are reduced using crystal symmetry. The nonlinear wave equation is solved in the parametric approximation when phase matching is not present. The boundary value problems for the fundamental entering the medium and for the second harmonic leaving are solved. The only significant null solution is found to be the Brewster's angle extinction. Some numerical exploration of the exact effects of absorption at both fundamental and second harmonic are made. They are found to be small and are negligible for qualitative purposes unless the imaginary part of the dielectric constant exceeds the real part. Variation of intensity with crystal orientation is described in detail. The occurrence of phase matching as a null solution of the wave equation is seen to offer a natural approach to the problem of obtaining the geometry of phase matching for the most general optical medium.

Security Classification

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Nonlinear optics						
Optical 2nd harmonic generation						
Nonlinear reflection from absorptive crystal						
Parametric approximation						
Phase match excluded						

DD FORM 1 NOV 65 1473 (BACK)
(PAGE 2)

Security Classification